Investigating the relationship between magnetization and the local structure in $La_{1-x}Ca_xMnO_3$; a probe of the magnetization process

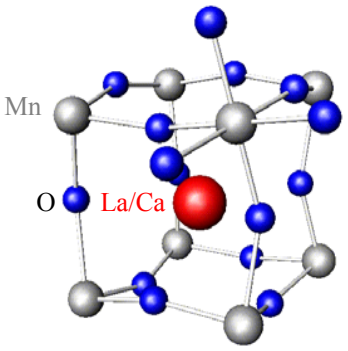
Frank Bridges, UCSC, DMR-0301971

Introduction

The "colossal" magnetoresistive (CMR) manganites, $La_{1-x}Ca_xMnO_3$ (x =0.2-0.5), experience polaron-induced local distortions near the ferromagnetic transition temperature T_c. Near T_c these distortions can be reduced by either lowering the temperature or increasing the applied magnetic field, effectively increasing the sample magnetization. We argue that this distortion is more generally a universal function of the magnetization rather than extrinsic quantities, such as the temperature and applied field.

Data

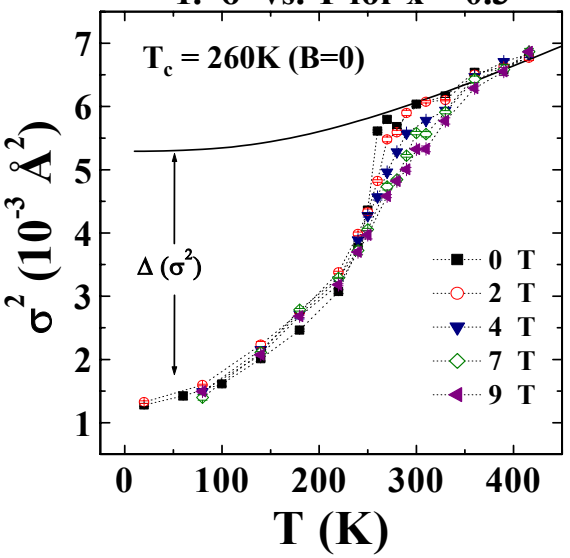
Mn K-edge EXAFS studies show that σ^2 (σ is the average width of the Mn-O bond length distribution, a measure of the local distortion) increases rapidly as T increases to T_c (as polarons form), and then changes slowly with T above T_c (see Fig 1). The amplitude of the Mn-O peak increases as the applied field increases from 0-9T near T_c (Fig. 2); this implies that σ^2 decreases; the reduction in σ^2 is observed from 250-350K (Fig. 1).



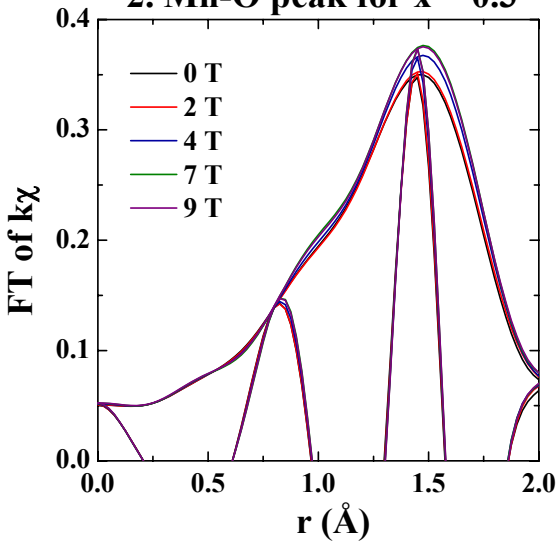
New Result

- Changes only occur near Tc. Below 150K, sample is fully magnetized, but distortions associated with polarons are still present. These distortions essentially disappear below 10K.
- Change in distortions ($\Delta\sigma^2$) depend only on M, irrespective of whether B or T is varied to change M.

1. σ^2 vs. T for x = 0.3



2. Mn-O peak for x = 0.3



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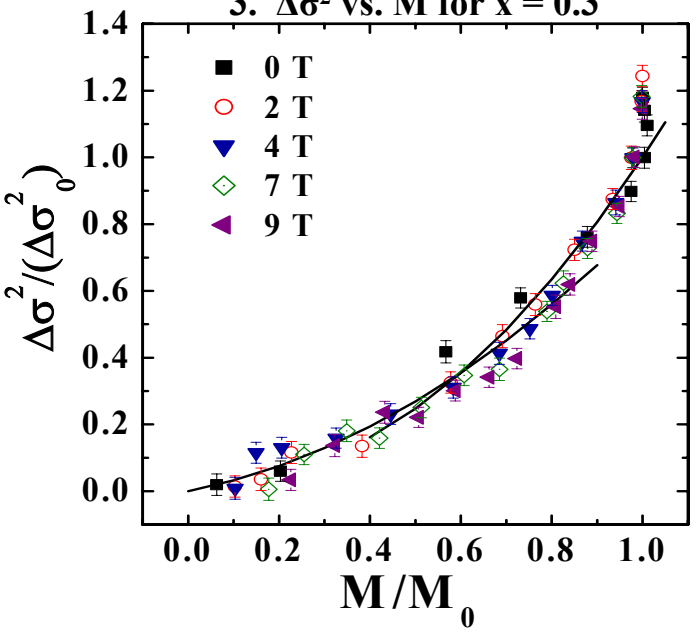
Model

- Fig 3 shows that $\Delta\sigma^2$ is a universal function of M; data for different fields overlap when plotted as a function of M.
- Distortions associated with polarons are removed as the sample becomes magnetized; however, these distortions first change slowly with magnetization at low M and then faster at high M, (above $M/M_0 \sim 0.6$ (2x) for this sample); lines show models.
- Suggests that sample becomes magnetized in pairs, i.e. an undistorted ("Mn^{+4}") and a distorted ("Mn^{+3}") site, until all undistorted sites are "used up". See Fig 4. The distorted "Mn^{+3}" becomes undistorted (pink site) when magnetized. Above $M/M_0 \sim 0.6$, a further magnetization of the sample requires that two distorted "Mn^{+3}" sites become undistorted, i.e. two red sites in Fig. 4 become pink sites. Thus the distortions removed increase more rapidly with M (above $M/M_0 = 0.6$).

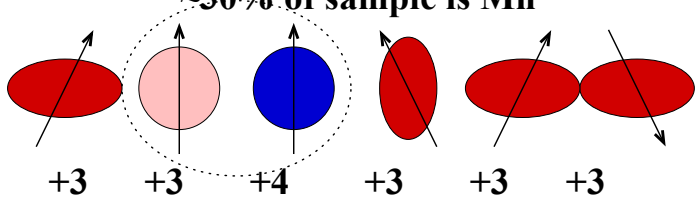
Broader implications

- A new model for the development of magnetization in the sample suggests that magnetic clusters initiate near "Mn^{+4}" sites and spread in a filamentary fashion throughout the sample via linked pairs.

3. $\Delta\sigma^2$ vs. M for x = 0.3



4. Model for magnetization process – ~30% of sample is Mn^{+4}



The pink Mn (+3 site) becomes undistorted when it becomes magnetized next to a Mn^{+4} site. The sample magnetization develops in filamentary clusters.

Educational

- Lisa Downward (Graduate student) worked on the EXAFS studies of CMR manganites as part of her PhD thesis.
- Lisa and several undergraduate students were trained to use the XAFS technique and run experiments at a Synchrotron source.